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Report Title

Final Progress Report:
Time-resolved scanning electron microscopy

ABSTRACT

The project explores the combination of ultrashort pulsed laser technology with electron microscopy. The objective is to build an electron microscope with a pulsed electron beam, in order to observe nanoscale structures with ultrafast time-resolution. The pulsed electron beam is obtained by rapidly switching the electron emission of a field emission tip using the AC electric field arising from exposure to the intense electromagnetic radiation emanating from an ultrashort pulsed laser. Space-charge interactions between electrons within the electron beam are minimized by constraining the number of electrons per pulse to less than about 10. The following operating parameters are targeted:
Spatial resolution: 2 nm; Temporal resolution: 3 ps
Ultrafast-pulsed electron microscopy could open up the wide and important field of dynamical processes in materials that are of nanotechnological importance.

List of papers submitted or published that acknowledge ARO support during this reporting period. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

"Megavolt electron beams for ultrafast time-resolved electron diffraction," F. M. Rudakov, J. B. Hastings, D. H. Dowell, J. F. Schmerge, and P. M. Weber, In "Shock Compression of Condensed Matter – 2005," ed. M. D. Furnish, M. Elert, T. P. RUssel, and C. T. White, American Institute of Physics (2006).

"Ultrafast Electron Microscopy in Materials Science, Biology, and Chemistry," Wayne E. King, Geoffrey H. Campbell, Alan Frank, Bryan Reed, John Schmerge, Bradley J. Siwick, Brent C. Stuart, and Peter M. Weber, Journal of Applied Physics, 97, 111101 (2005).

"Experimental and theoretical studies of pump-probe electron diffraction: time-dependent and state-specific signatures in small cyclic molecules," Peter M. Weber, Ray C. Dudek, Seol Ryu, and Richard M. Stratt, in "Femtochemistry and Femtobiology: Ultrafast Events in Molecular Science," Eds. M. Martin and J. T. Hynes, Elsevier, p. 19, (2004).

"Probing reaction dynamics with Rydberg states: The ring opening reaction of 1, 3-cyclohexadiene," N. Kuthirummal and P. M. Weber, in "Femtochemistry and Femtobiology: Ultrafast Events in Molecular Science," Eds. M. Martin and J. T. Hynes, Elsevier, p. 37, (2004).

"Centering of ultrafast time-resolved pump-probe electron diffraction patterns" J. D. Cardoza, R. C. Dudek, R. J. Mawhorter, and P. M. Weber. Chemical Physics, 299, 307 – 312, (2004).

Number of Papers published in peer-reviewed journals: 5.00

(b) Papers published in non-peer-reviewed journals or in conference proceedings (N/A for none)

Number of Papers published in non peer-reviewed journals: 0.00

(c) Presentations

Lawrence Livermore National Laboratory, July 22 2005: “MeV-UED: A Start!”

11th European Symposium on Gas Electron Diffraction, Blaubeuren, Germany, June 27, 2005: “Pump-probe diffraction with very slow and very fast electrons.”

88th Canadian Chemistry Conference, Saskatoon, CA, May 29, 2005: “Excited state structure and dynamics as probed with very slow and very fast electrons.”

MIT, May 03, 2005: “New Techniques for Time-Dependent Structure Characterization.”

Talks held by collaborators:

Femtochemistry VII, Washington D.C. July 17-22 2005 “Ultrafast Dynamics in Rydberg States of Tertiary Amines” M.P. Minitti, J.L. Gosselin, T.I. Solling and P.M. Weber

60th annual Ohio St. University International Symposium on Molecular Spectroscopy, The Ohio State University June 20-24 2005 “Ultrafast Dynamics in Rydberg States of Aliphatic Amines” J.L Gosselin, M.P. Minitti, T.I. Solling and P.M. Weber

2004 Focused Interest Group, Materials Research in an Aberration Free Environment, Pre-Congress Meeting, July 31 and August 1, 2004, Savannah, Georgia: “Toward Ultrafast Electron Microscopy” Wayne E. King, G. H. Campbell, Alan Frank, Bryan Reed, John Schmerge, Brad Siwick, Brent Stuart, and Peter Weber; Talk held by Wayne E. King, LLNL.

Number of Presentations: 7.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts): 0

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts): 0

(d) Manuscripts

“Ultrafast time-resolved electron diffraction with megavolt electron beams,” J. B. Hastings, F. M. Rudakov, D. H. Dowell, J. F. Schmerge, J. Cardoza, J.M. Castro, S.M. Gierman, H. Loos, and P. M. Weber, submitted for publication, Appl. Phys. Lett.

Number of Manuscripts: 1.00

Number of Inventions:

Graduate Students

NAME	PERCENT_SUPPORTED	
Fedor Rudakov	1.00	No
FTE Equivalent:	**1.00**	
Total Number:	**1**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
Peter M. Weber	0.08	No
FTE Equivalent:	**0.08**	
Total Number:	**1**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Personnel receiving masters degrees

NAME
Total Number:

Names of personnel receiving PHDs

NAME	
Wei Cheng	No
Total Number:	**1**

Names of other research staff

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Sub Contractors (DD882)

Inventions (DD882)

Review of Past Work

In the current funding period, we have explored the generation of ultrashort electron beams suitable for electron microscopy. Attention was paid to the particular requirement for this situation, such as excellent brightness, spatial focusing, and minimization of space-charge interactions. In order to obtain electron pulses suited for electron microscopy, we are exploring an approach that radically differs from other methods.

Usually, the fast-pulsed electron beam is generated using a train of ultrashort laser pulses. Electrons are ejected from a metallic photocathode in either a single-photon or multi-photon emission process. Figure 1 illustrates this approach. Once the electrons leave the surface, a DC field accelerates them to the target.

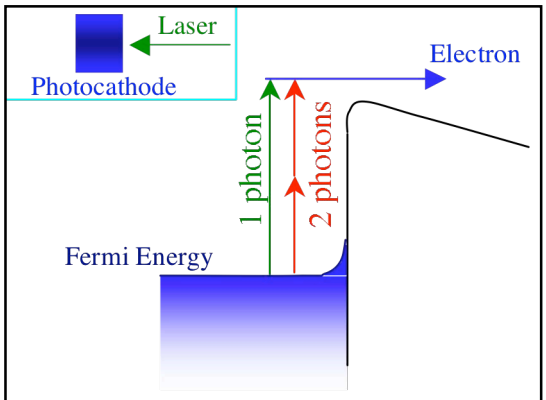

Figure 1: One-photon and two-photon ejection of electrons from a metallic surface by the photoelectric effect.

This approach using the photoelectric effect is used in almost all pulsed electron experiments today. In particular, this includes our past work on time-resolved electron diffraction, but also work done at large national facilities where pulsed electron beams with Megavolt energies are created. The advantage of this approach is experimental simplicity. The disadvantage is that right after the electrons are ejected, they move slowly. At that point, space charge interactions between electrons within a pulse greatly increase the duration of the electron pulse. While this effect can be countered in relativistic electron diffraction experiments using radiofrequency compression techniques, for the purpose of electron microscopy this is a serious drawback.

Our new approach is dramatically different. We employ the ultrafast, oscillating AC electric field component of the laser pulses, aided by a large static field similar to those found in field emission microscopes, to eject the electrons. The idea is illustrated in figure 2. The applied DC voltage provides an electric field that is large, but not quite large enough to allow electrons to tunnel. Thus, there is no electron ejection at this point. When the ultrashort laser pulse impinges on the tip, however, the high frequency electric fields from the electromagnetic radiation add

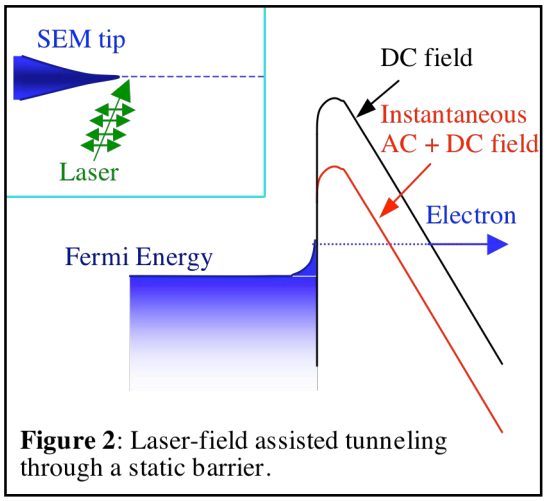

Figure 2: Laser-field assisted tunneling through a static barrier.

to the DC field. When the direction of the AC field adds to the DC field, the barrier for electron tunneling from the surface is lowered. If experimental conditions are right, this allows the tunneling of the electrons through the barrier.

The advantage of our new scheme is that electrons can only be ejected for the duration of the laser pulse. Given the nonlinear nature of the effect, one can also adjust the parameters such that only the central portion of the laser pulse leads to electron emission. Most importantly, the combined large DC and AC fields immediately accelerate the electrons that leave the surface. As a result, they spend much less time near the photocathode, and therefore there is much less time for space-charge broadening of the electron pulse. Finally, since the electrons are ejected from the tip of the field emitter, their spatial characteristics, in particular its brightness, are excellent.

In order to test the ultrafast pulsed electron ejection mechanism, we have performed experiments with a test apparatus (see figure 3). A laser pulse from a femtosecond pulsed Titanium Sapphire laser with a wavelength of 800 nm and a pulse duration of 80 fs, is focused onto the tip of a needle with 10 μm radius. Initially we tried commercial tips for field emitter microscopes, but we found them too unstable. Later experiments employed a tungsten tip that we produced ourselves (see photograph in inset of figure 3).

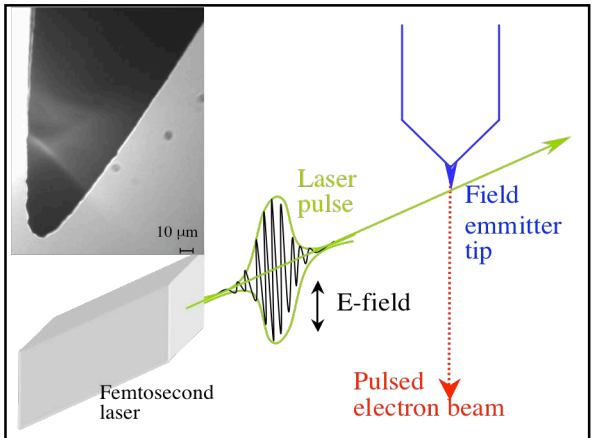

Figure 3: Generation of a pulsed electron beam using an ultrashort pulsed laser.

In our test experiments we found that electron ejection from this device is facile, and that large electron currents can easily be obtained. Even with modest DC fields and easily obtainable laser powers, currents in the range of tens of nanoamperes were obtained. This greatly exceeds the currents usually achieved with planar photocathode devices. Figure 4 shows the dependence of the current on the DC acceleration voltage, for different laser powers. It is clear that the DC field has a significant role in the electron ejection, and that without such a field the electron emission ceases. At the same time, it is also clear that the field emission by the static field alone does not suffice to produce the electron beam in our experimental conditions.

The dependence of the electron current on the intensity of the femtosecond laser beam is shown in figure 5. Again, it is apparent that both the laser field and the DC field need to be present to create the electron beam, supporting the suggestion that we observe the electrons that are ejected by the desired combination of DC field and the laser field.

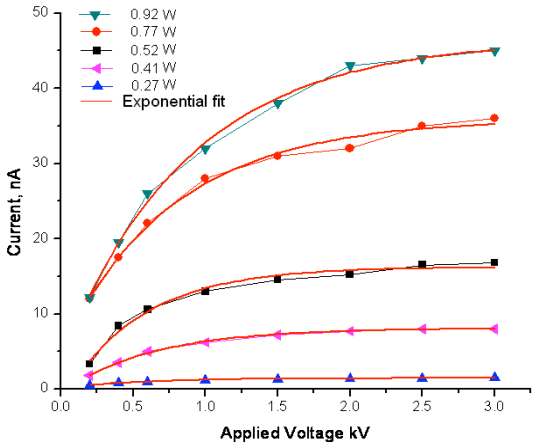

Figure 4: Electron beam currents obtained for different laser powers (as indicated in legend), as a function of DC acceleration voltage. The solid lines are fits using an exponential rise.

Interestingly, the observed electron current appears to saturate at high laser power. The source of this saturation is not yet clear. One suggestion is that at large electron currents, there may be additional fields between the just-ejected electrons and the mirror charges left behind in the emitter tip. At 30 nA current, a current that is readily obtained for the higher laser powers, each laser pulse generates about 2500 electrons at the tip. The interaction of that charge cloud with the positive mirror charge left behind could pull electrons back into the emitter, thus retarding the electron current that can be obtained.

Several further observations strengthen the assignment of the observed current to the dual DC/AC field emission. For example, we find that for the same average laser power, but cw laser exposure, we find no electron emission. This is easily understood because the AC field of the cw laser is small. The result is important because it eliminates a thermal mechanism of electron ejection. Separately, we found that when the polarization of the laser field is perpendicular to the electron beam axis, we observe no current as well. Clearly, it requires the addition of the collinear fields to achieve the desired effect. This indicates that it is not a traditional multi-photon ejection of the electrons that is at play in generating the electron beam.

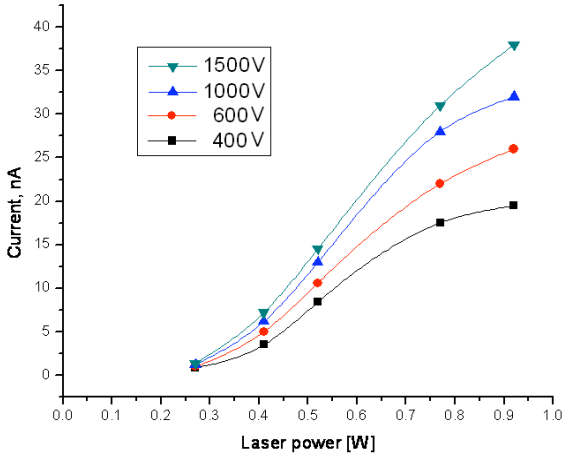

Figure 5: Electron beam currents obtained from combined DC and laser field emission, as a function of the laser power, for different DC voltages (as indicated in legend).

Summary of past work

We have developed a novel electron ejection scheme where the fields from an applied DC voltage and the AC field from an ultrafast laser pulse combine to eject electrons from the tip of a needle. Using a test apparatus, we have generated large electron currents with this new scheme. All tests support the two-fold origin of the electron ejection.

The new technique to generate ultrafast pulsed electron beams is ideally suited for electron microscopy. It generates large electron currents while preserving the high brightness that characterizes field emitters. The demands on the laser power are modest, and can readily be met with commercial off-the-shelf laser hardware.